Indian J. Phys. **49**, 596–602 (1975)

Thermally stimulated discharge currents in bees' wax electrets

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(*Received* 10 *December* 1974, *revised* 14 *April* 1975)

Thermally stimulated discharge currents have been obtained from Bees' wax electrets prepared at various polarising voltages in the temperature range 52°C to 62°C. Total heterocharge released on depolarisation is not linear with applied polarising field indicating non-uniform volume polarisation Hence besides dipole orientation, the charge injection from the electrodes and migration of ions with microscopic displacement with trapping with subsequent space charge build up is the proposed model

1. INTRODUCTION

An electret is a dielectric slab exhibiting a permanent electric field. Heaviside (1919) was the first who got the idea of electret and Eguchi (1925) made the first electret by using carnauba wax The most of the experimental work on electret as reported in the litreature so far are confined to the study of the nature of surface charges, their magnitude and period of retention and effects of polarising field and temperature on the nature of surface charges. But very little work has been mentioned about the factors responsible for the formation of electrets Investigations of the electret state opens a new chaptor for studying the mechanism of polarisation of dielectrics. Theoretical explanations regarding formation of electret are still inconclusive. Gemant (1935) proposed that the heterocharge on the electret is due to displacement of ions and dipole orientation inside a dielectric. Important contributions were made by Gross (1949) and confirmed by Wiseman & Feaster (1957). According to them heterocharge is formed by the process of charge absorption in dielectrics, while homocharge is due to break down at the dielectric-electrode interface. The heterocharge is an internal volume effect, the nature of which remains in doubt. A non-uniform volume polarisation can be produced by space charge build up associated with the migration of ions within the dielectric over microscopic distances or charge injection from the electrodes. A uniform volume polarisation is produced by dipole orientations or migration of charge over microscopic distances with trapping or both

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Recently Perlman (1971) and Campos (1971) have used the technique of thermally stimulated currents for analysing the electret effect and for studying of trapping parameters in electret. They hold the view that polarisation is uniform in carnauba wax electrets and charges suffer microscopic displacement during polarisation and are trapped. This paper reports on our work on the depolarisation of the thermoelectret state of bees' wax. It is mainly concerned with the study of the thermal release of charges believed to arise from the trapping of electrons and holes injected by electrodes along with migration of charges over microscopic distances and dipole reorientation. The results throw more light on the main points of dispute in electret studies, that is, whether a uniform volume polarisation or a space charge mechanism or a combination of two is responsible for the electret state formation.

2. Experimental

Thermoelectrets were prepared from commercially obtained bees' wax (imported). The bees' wax sample contained a number of ingradients and hence it did not have a sharp melting point. However, the sample melted completely between 60°C to 61°C. The principle of measurement employed in the present work was that of two parallel plate electrodes capacitor. Both the electrodes were made of aluminium of size three cm in diameter. Lower electrode was fixed in small copper basin while upper electrode was made to move vertically on screw arrangement. The separation between the electrodes was kept one mm The copper basin was placed on insulated base made of asbestos. This assembly was kept in thermostat (model Hoppler NBE) and an electric field was applied by high voltage power supply (model HV211, Electronic Corporation of India). A positive polarising voltage was applied to the upper electrode while the lower electrode was grounded. The temperature was raised and at the fixed temperature the sample was left for three hours and it was then cooled to room temperature in the presence of electric field. It took nearly three hours to reach the room temperature. After taking out the sample from thermostat, it was kept in desiccator in short circuit condition to remove all the stray charges developed due to the close contact between the metallic electrodes and the dielectric. After 15 minutes the sample was again placed in thermostat which could be heated from room temperature to 70°C. The samples were depolarised at a linear heating rate of 1°C per five minutes. The depolarisation current was recorded with upper electrode connected to an electrometer amplifier (model EA810, Electronic Corporation of India) with lower electrode grounded. The thermal current spectrum was recorded from the room temperature to above the polarising temperature that is nearly 70°C.

Different sets of readings were taken on depolarising the electret made at the temperatures 52°C to 62°C at different polarising voltages (800 volts, 1000 volts, 1200 volts and 1400 volts per mm thickness of the sample).

3. Theory

The potential developed on the electrodes of a shorted capacitor are due to induction by fields which originate from charges residing in the dielectric after polarisation. Observation of the induced charge of the electrodes therefore gives information about the nature of the charges present in the dielectric at different temperatures.

The volume polarisation leads to the appearance of the surface charges, $S = -PA$, where S = surface charge, P = polarisation and A = area of the electrodes. The polarity of S is contrary to that of the corresponding electrode during polarisation, thus accordingly S is heterocharge.

When the applied voltage is sufficiently high and the temperature well above room temperature or even when very high voltage is applied at room temperature, the transfer of charge takes place from electrodes to the dielectric. Thus the electrode which was connected with the positive pole of the high voltage unit carries a positive induced charge and the other electrode negative induced charge hence known as homocharges. Thus it is attributed that at the moment of short circuit the dielectric carries two types of surface charges : (i) heterocharges S, and (ii) homocharges f. Thus the total induced charge on the electrode is given by

$$q = -(S+f), \qquad \ldots \quad (1)$$

where the sign of f is contrary to that of S

A quantitative idea about the charges is obtained by measurement of total external current $J(t)$ which is given by equation

$$J(t) = i(t) + \frac{dq}{dt}, \qquad (2)$$

where $i(t)$ is conduction current across the dielectric electrode interface which measures the decay of homocharge and dq/dt is the displacement current which measures the decay of heterocharge Heterocharge $S(t)$ may be expressed as

$$S(t) = \int J(t)dt, \qquad \ldots \quad (3)$$

where external current $J(t)$ mainly originates due to decay of heterocharge.

The discharge current $J(t)$ in the external circuit is due to the decay of volume polarisation. Bucci (1964) on dipolar theory for ionic thermocurrent has given the expression for the glow peak of the discharge current $J(t)$ during the heating of the sample at constant rate

$$J(t) = \left(\frac{N\mu^2 E_p}{3KT_p\tau_0}\right) x \exp\left[-\frac{U}{KT} - \frac{1}{\beta\tau_0}\int_{T_0}^{T} \exp\left(-\frac{U}{KT}\right) dT\right], \quad \ldots \quad (4)$$

where μ = dipole moment, N = dipole concentration, K = Boltzmann's constant T_p and E_p = polarising temperature and field, β = rate of heating and U = activation energy and τ_0 = a constant given by formula $\tau = \tau_0 \exp(U/KT)$, where τ is the relaxation time at the temperature T. Perlman (1971) and Pillai (1973) used this technique and found that eq. (4) for the discharge current also holds true in the case of microscopic charge displacement with trapping mechanism which is responsible for the heterocharge From eq. (4) it is seen that Q which is total charge released to the external circuit should be independent of the thickness of the sample provided that the polarising field E_p and temperature T_p are kept constant.

The total charge released is given by

$$Q = \int J(t)dt. \qquad \ldots \quad (5)$$

On differentiating the eq. (4), the temperature T_m at which maximum current occurs $(dJ/dt = 0)$ can be written as

$$\tau_0 = \frac{KT_m^2}{\beta U \exp\left(\frac{U}{KT}\right)}. \qquad \ldots \quad (6)$$

Thus T_m is independent of both E_p and T_p but it is dependent on β.

4. Results

After preparation of electret of bees' wax sample under different polarising fields (800 volts, 1000 volts, 1200 volts and 1400 volts) at different temperatures 52°C, 56°C, 58°C, 60°C and 62°C), the thermal currents spectra were recorded. The curves were drawn for different polarising voltages keeping the polarising temperature constant. Figure 1 is one of the representative curves of thermal current spectra for different polarising voltages at the polarising temperature 58°C.

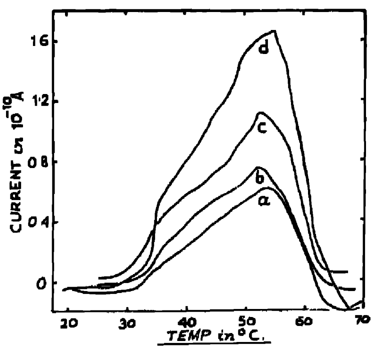

Fig. 1. Current vs temperature curves. Polarising temperature = 58 °C. The curves (a), (b), (c) and (d) corresponding to E_p = 800, volts, 1000 volts, 1200 volts and 1400 volts per mm thickness of the sample respectively.

Similarly to study the effect of polarising temperature, the curve for thermal current spectra for different temperatures at constant field were drawn. Figure 2 is one of the representative curves of thermal current spectra for different polarising temperatures at 1400 volts per mm thickness of the sample.

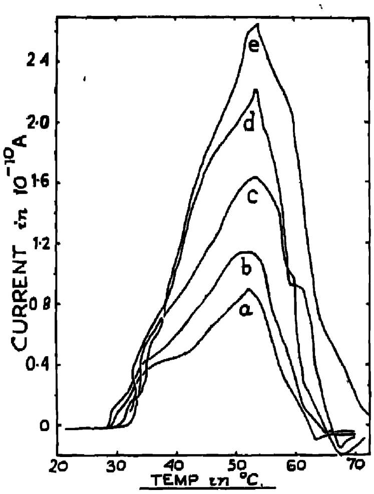

Fig. 2. Current vs temperature curves. Polarising field 1400 volts/mm The curves (a), (b), (c), (d) and (e) correspond to T_p = 52 °C, 56 °C, 58 °C, 60 °C and 62 °C respectively

It is seen that the peak position in both figures 1 and 2 is at almost a constant temperature T_m with a little variation The magnitudes of peaks in both the above cases gradually increases with the increase of polarising voltages and temperatures.

In both figures 1 and 2, it is seen that thermal currents in the beginning gives the negative value up to temperatures nearly 30°C to 32°C then positive value, showing peaks at nearly 52°C to 55°C Currents sharply decrease to zero at nearly 61°C to 65°C and then after pass to negative value.

The areas of the positive portions of the curves of thermal current spectra for different polarising voltages and temperatures were calculated. Figure 3 is a plot of area of the positive portion of the curve versus polarising voltages. It is seen that the relationship is not linear indicating non-uniformity in volume polarisation.

5. Discussion

When a capacitor containing bees' wax is charged at high voltage and temperature and then cooled to room temperature in the presence of the electric field, homo and hetero charges are *frozen in* and retained in the dielectric as long as temperature is low. After short circuiting the sample for some time and then on reheating without any external voltage, the frozen in charges are released and discharge current $J(t)$ as given by eq. (2) is obtained. In the lower temperature range up to nearly 32°C, the displacement current dq/dt is negligible and the discharge current is mostly due to the decay of homocharge in the form of conduction current between the dielectric—electrode interface. In the temperature range say from 32°C to 60°C or 65°C, the discharge current is positive and passes through a peak. This current seems entirely due to the decay of the heterocharge originating from volume polarisation. Beyond 60°C or 65°C the current again changes the direction and small conduction current due to homocharge persists. So it is concluded that first there is homocharge then heterocharge and again homocharge on the surface of the dielectric in contact with upper electrode which was connected with positive voltage. The heterocharge $S(r)$ as the integral over $J(t)$ was calculated from the area of the positive portion of the curve.

Eq. (4) is true either for dipole orientation mechanism involved or microscopic charge displacement with trapping responsible for heterocharge in the electret state in the material Eq. (6) suggests that peak temperature T_n should be independent of polarising field E_p and polarising temperature T_n. It is seen from figures 1 and 2 that there is little variation in peak temperature T_n which may be due to slight change in heating rate β So eqs (4) and (6) are in complete agreement with results. But Gerson (1955) ruled out a pure dipole orientation mechanism in electrets because even complete dipole alignment is insufficient to produce the observed charge and Perlman (1971) calculated the number of dipoles and mentioned that too many dipoles had to be aligned to produce the observed effects. Moreover, heterocharge has been also observed in non-polar substances. Thus it is felt that the polarisation may partly result from the orientation of the dipoles and partly from microscopic displacement of charge carriers trapped in bulk of the material.

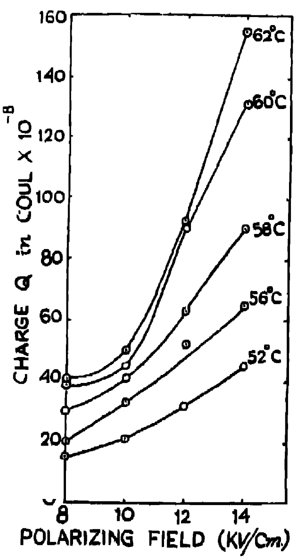

Fig. 3. Total heterocharge released to external circuit vs polarising fields.

Further, from the plot of figure 3, it is seen that total heterocharge released is not linear with the applied polarising field hence a non-uniform volume polarisation may be predicted which suggests that along with migration of charge over microscopic distances with trapping and dipole orientation, there is space charge build up associated with the migration of ions within the material over microscopic distances or charge injection from the electrodes with trapping at different sites. As pointed out by the authors (1973, 1974) in the study of the conduction mechanism in bees' wax, the boundaries between crystalline and amorphous regions provide additional trapping sites for charge carriers.

6. Conclusions

The study of depolarisation of bees' wax thermoelectret has helped in understanding the mechanism responsible for charge carriers *frozen in* in the electret From the above discussion it is concluded that

(1) Some time after short circuit and on reheating there is homocharge then heterocharge and lastly homocharge which persists

(2) Decay of homocharge is in the form of conduction current between dielectric—electrode interface and decay of heterocharge is in the form of displacement current.

(3) Heterocharge originates from the volume polarisation. This polarisation is partly due to orientation of dipoles and partly from microscopic displacement of charge carriers trapped in the volume of the material.

(4) There is non-uniform volume polarisation hence space charge build up is expected.

Acknowledgment

The authors are thankful to the Department of Industries and Technical Education, Bihar Government for providing financial support to one of the authors (S.N.P.). We are specially indebted to Dr. L. S Singh, Head of the Department of Physics, Patna University for providing all facilities and kind encouragement in the work.

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